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Enhanced two-demensional precipitation of excess As in LT-GaAs delta-doped with Sb

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Abstract. We have used, for the first time, isovalent delta-doping with antimony instead of indium to produce two-dimensional sheets of arsenic nano-clusters in GaAs films grown by molecular beam epitaxy (MBE) at low (200°C) temperature. The precipitation kinetics at Sb delta-layers is found to be enhanced as compared to that for In delta-doping. It provides an opportunity to improve the cluster spatial ordering. In addition, at early precipitation stages, the microstructure, morphology and orientation relationship of the clusters attached to Sb delta-layers are found to differ from those conventional for As clusters in LT-GaAs films including As clusters at the In delta-layers.

1 Introduction

GaAs grown by MBE at low substrate temperatures of 200–250°C (LT-GaAs) is a very attractive material for microwave and optoelectronic applications [1–3]. Being nonstoichiometric in an as-grown state, it contains an extremely high concentration (10^{20} cm^{-3}) of intrinsic point defects of which the main are excess As atoms in the forms of As antisites and As interstitials. Upon subsequent annealing, the excess As precipitates in nm-size clusters buried in GaAs matrix, and the material exhibits a high electrical resistivity, high carrier mobility and very short lifetime of nonequilibrium charge carriers. Since the As clusters play a crucial role in the electrical and optical properties of LT-GaAs, it is of importance to control the density and spatial distribution of As cluster arrays. The As cluster density has been shown to vary from layer to layer in LT-AlGaAs/GaAs [4] and LT-InGaAs/GaAs [5] heterostructures. An accumulation and depletion of As clusters can be also produced [6] by nonuniform incorporation of an impurity. When Si or In is inserted in a LT-GaAs film as delta-layers, two-dimensional As cluster sheets have been demonstrated [7-8] to appear. Before, we succeeded [9-11] in obtaining two-dimensional precipitation of As clusters in LT-GaAs films uniformly doped with Si donors, Be acceptors or undoped by means of additional delta-doping with In that is an isovalent impurity replacing Ga in group III sublattice. In this work, we use, for the firs time, delta-doping with antimony, i.e. group V element, and show an enhanced two-dimensional precipitation of excess As as compared with the case of indium delta-doping.

2 Experimental

The LT-GaAs films used in this work were grown by MBE at a substrate temperature of 200°C on undoped semi-insulating 2-inch GaAs(001) substrates in a dual-chamber system "Katun". The films contained 80 nm spaced Sb delta-layers which were introduced in the GaAs matrix by using the antimony beam while the arsenic beam was interrupted. The Sb deposit in each delta-layer was equivalent approximately to 1 monolayer (ML). Similar LT-GaAs films delta-doped with indium were also grown at the same growth temperature and As/Ga beam ratio. The growth procedure is described in more detail elsewhere [12]. The grown samples were cut into few parts of which one was kept as-grown and the others were annealed under As overpressure at 500, 600 or 700°C for 10, 30 or 60 min.

Optical absorption measurements in the near-infrared (0.8–1.2 mm) region (NIRA), conventional (TEM) and high-resolution (HREM) electron microscopies were applied to study the samples. Philips EM 420 and JEOL JEM 4000EX microscopes were exploited. Cross-sectional TEM specimens were prepared by a conventional Ar^+ ion-milling procedure as well as by cleaving technique.

3 Results and discussion

Cross-sectional TEM of the as-grown LT-GaAs films revealed thin layers located just at the positions expected for Sb or In delta-layers from the growth procedure. NIRA spectra recorded at room temperature showed an increased absorption, conventionally attributed to As antisites, in all the as-grown samples. The As excess was estimated from these spectra to be 0.5 at.% for both Sb delta-doped and In delta-doped LT-GaAs films. The equal contents of excess As in both kinds of the samples idicate that isovalent Sb doping does not supress the extra As incorporation during the film growth.

The microstructure of the Sb delta-layers was studied by HREM. Observations of the Sb delta-layers in the as-grown samples by lattice imaging revealed Sb to be dispersed within 4–5 ML that is similar to the case of In delta-doping [13] and results from some surface roughness of the film growing at a temperature as low as 200°C. Despite

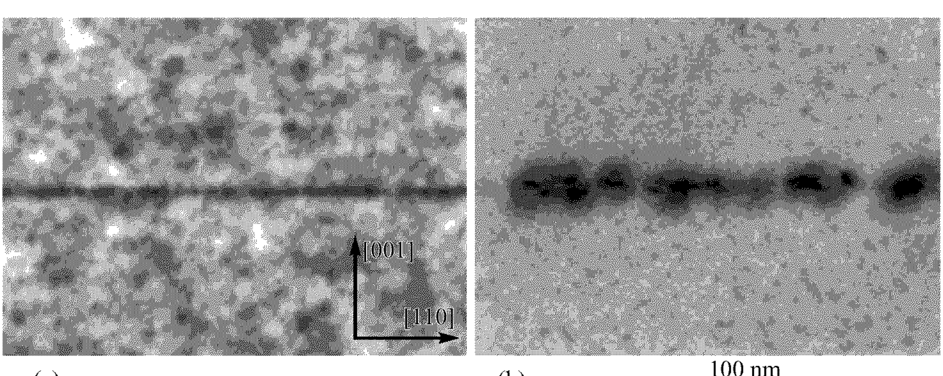

Fig 1. Dark-field 002 cross-sectional images of LT-GaAs films delta-doped with In (a) and Sb (b) and annealed at 500°C for 10 min. An essential difference in cluster accumulation at the In and Sb delta-layers is clearly seen.

this broadening the delta-layers accumulate clusters which are forming upon annealing, whereby the precipitation kinetics is found to differ for Sb delta-doping and In delta-doping. Fig. 1 presents TEM micrographs taken from the In or Sb delta-doped films grown and annealed under the same conditions. As can be seen, the clusters attached to the Sb delta-layers are essentially bigger and, in addition, provide stronger strain-induced contrast than those at In delta-layers. The lateral size of the clusters at the Sb delta-layers is up to 7 nm while it reaches only 3 nm for In delta-doping. In contrary, clusters dispersed in-between the delta-layers occur to be smaller in the case of Sb doping.

Annealing at higher temperatures (600, 700°C) led to an increase in size of the clusters attached to Sb delta-layers, that is illustrated by Fig. 2. The cluster size has been estimated to be 13 nm for an annealing temperature of 600°C and 19 nm for an annealing temperature of 700°C. At the same time, the estimated density of the clusters attached to the Sb delta-layers reduced from $6 \times 10^{10}\,\mathrm{cm}^{-2}$ down to $3 \times 10^{10}\,\mathrm{cm}^{-2}$. In parallel, the size of small clusters suspended in-between the Sb delta-layers increased from 2 nm up to 5 nm when elevating annealing temperature from 500 to 700°C. The cluster density in three-dimensional array in-between the Sb delta-layers reduced one order of magnitude: from $1.5 \times 10^{17}\,\mathrm{cm}^{-3}$ for an annealing temperature of 500°C down to $1.6 \times 10^{16}\,\mathrm{cm}^{-3}$ for 700°C.

The cluster microstructure and morphology for high annealing temperatures (600–700°C) have been observed to be similar to those conventional for As clusters in LT-GaAs films including clusters attached to In delta-layers. Fig. 3a shows a high-resolution image of such a cluster exhibiting moire fringes running nearly along (111) GaAs matrix planes. However, at the initial growth stages the microstructure, morphology and orientation relationship of the clusters attached to Sb delta-layers have been found to differ from conventional ones. It is demonstrated by Fig. 3b where a high-resolution image of the cluster attached to Sb delta-layer in LT-GaAs film annealed at 500°C is presented. On our opinion, two reasons may contribute to this phenomena. First, along with excess arsenic, antimony can take part at the cluster nucleation an growth. Second, the strain distribution around the Sb delta-layers can be different from that around the In delta-layers. The changed strain is indicated by a strong strain contrast at the dark-field image (Fig. 2) of the clusters at the Sb delta-layers.

4 Summary

So, the cluster accumulation happens more effectively in the films delta-doped with Sb as compared with those delta-doped with In. It provides an opportunity to improve the cluster spatial ordering. In addition, at early precipitation stages, the microstructure, morphology and orientation relationship of the clusters attached to Sb delta-layers are found to differ from those conventional for As clusters in LT-GaAs films including As clusters at the In delta-layers.

Acknowledgments

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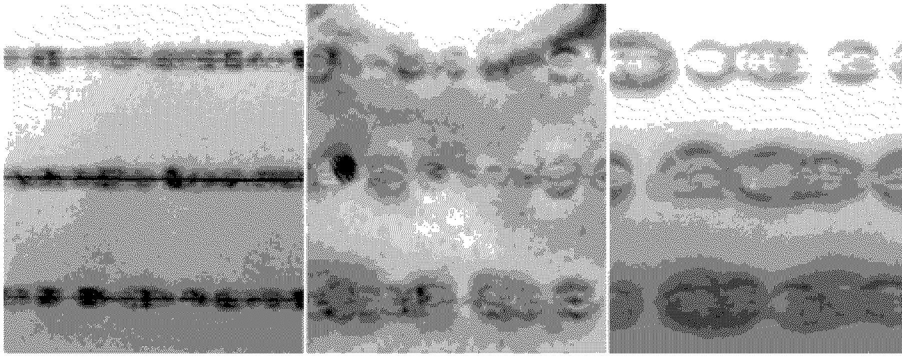
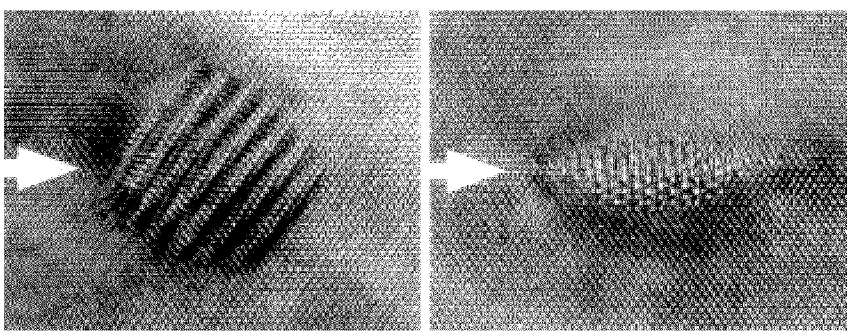


(a) 100 nm (b) (c)

Fig 2. Dark-field 002 cross-sectional images showing the clusters growing at the Sb delta-layers when elevating annealing temperature from 500 (a) to 600 (b) and to 700°C (c). The annealing time is 10 min in each case.



(a) (b) 10 nm

Fig 3. High-resolution images along [010] of the clusters attached to the Sb delta-layer in LT-GaAs films annealed for 10 min at (a) 600°C and (b) 500°C. The position of the Sb delta-layer is arrowed.

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